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# Application of Silica Supported Aluminium Hydroxide as a Low Cost Green Catalyst for the One Pot MCR Sythesis via Thermal Method

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## ABSTRACT

The catalysis is the first and active side they are good yield with good reusability. The sequence of time. They are the time of reaction is smaller, it saves the energy and time consummation, another thing is the good catalyst is intelligent for the dynamic yield but are focused on the design the catalyst they have good effectiveness as well as good reusability with green chemistry concept. In today times much, research is being carried out on Nano particles green catalysis we have eco-friendly make an endeavoured for designing new catalyst from waste aluminium foil to new catalyst. So our catalyst is designed from waste aluminium foil and transformed into good and reusability catalyst. They are continuing produced the same yield up to 7-8 reaction cycle.

**Keywords:** ketone, Aromatic aldehydes & ammonium hydroxide hydrochloride, green path with help of silica supported catalyst.

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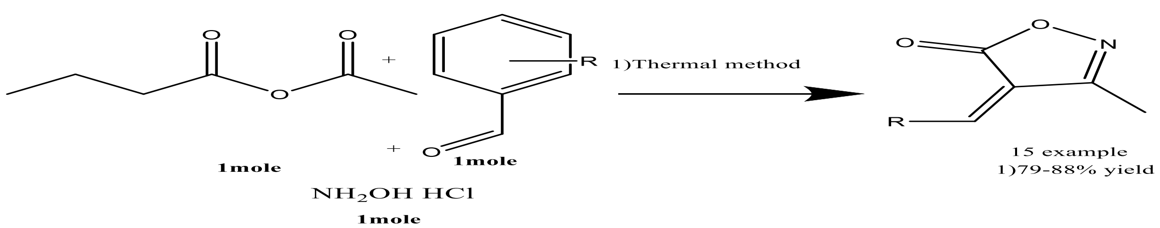
## Introduction

In modern life, waste management is the fastest flourishing discipline of research. The metal-based waste is the major hurdle for the world. We typically use aluminium foil for the protection of the over food from Bactria, but they have generated the enigma after the through external because they are extremely willing to the spared the fire site of garbage and they generate the position gas. The Aluminium is the second biggest metal are using in the current world, so we used the aluminium waste as a catalyst in our MCR reactions (2). we have carried out the preparation of mesoporous silica-aluminium according to of prepared mesoporous silica-aluminium hydroxide of Nasser h co-workers. photo degradation of methyl orange. (3) Because of our MCR synthesis base use as a catalyst for the reactions Na2S(5), DBCO(6), NBS(7), KHP(8), Citric acid(9), Ag/SiO2(10), Pyridine(11), boric acid(12) is economically not affordable catalyst so Rajesh and his co-workers are design the reaction by the starch solution used as a catalyst. they are outlined isoxazole-s-(4H)-one by a thermal method as well as microwave method. so we are altered and check the different catalyst for the environment favourable as well as expense-friendly so we make aluminium hydroxide from waste aluminium foil and then supported by the Silica they BET area and our silica supported aluminium hydroxide BET area so; they are more active than current catalyst and its reusability is 8 to 9 times they are not then another catalyst. Reaction time and the temperature is almost the same, but reusability of a catalyst is greater than another catalyst it's over the good site in the field of the catalyst.



## Experimental

## Material & Method:

### Material:

All chemicals are provide by HPLC lab chemicals. the TLC plate got from MERCK, Analysis supported by NFDD,RAJKOT(IR,NMR,MASS).

### Method for catalyst Preparation:

First our team collect the aluminium waste foil from food court and wash with water remove the food sample from foil then dry it cuts in small pieces dissolved in Waste acid (collect from industry) set Ph=2.0 of solution then adding the sodium hydroxide pallets and turns in to aluminium hydroxide white precipitation are formed its insoluble in water and other organic solvents. then precipitate are filter by vacuum filter and dry at 100*C in Hot air oven free fall white powder is formed then its docking with silica and 300*C in muffle furnace then collect it.

### Method Selection for Efficient path:

This activity for the selection of the efficient path for the reaction. first aldehyde(1mole),ethyal aceto acetate(1mole) & ammonium hydroxide hydrochloride mix well and starring for 2-3 min after completely mixing add the catalyst in assorted amount and legion temperature, and hind most fable path for the reaction. then reaction mass is dissolved in the ethanol and collect the catalyst for next reaction and re crystallization of product.dry the catalyst use for the next reaction. Take the TLC measured the $R_f$,

## Results & Discussion:

Ruling the Multi component reaction is confide on time and amount of catalyst, for the environment in addition to green path for the synthesis, the conformation of the final structure By IR,NMR & MASS analysis, the conformation of the catalyst with help of BET analysis for the check the surface and porosity of the silica supported catalyst. the porosity of the catalyst is change with respect to the reaction cycles, yield is decreases with reaction cycles. our team is carried out the with or without silica docking aluminium hydroxide its gave good yield and good efficiency in docking with silica there for over reaction is carried out work by the docking catalyst. this reaction is the mixture of ethanol and water as a solvent and also try to solvent free condition but its not gave tolerable result, for the check the result of catalyst (1:1:1)(ethyl acetoacetate: armomatic aldehyde, hydroxylamine hydrochloride) and add catalyst different amount like(0.1,0.2,0.3,..0.5) as per mole ratio as per reactant amount, then reaction take various temp like RT,45,90 degree celious then we saw the amount(0.5 equ.molar) of catalyst and temp(90-95'c).its more favourable condition for the preparation of 3-methyl-4-(4methylbenzylidene) isoxazol-5(4H)-one. we are found the electron rich group are gave good yield after the reactions and the approximately the catalyst are almost equivalent work up to 7 to 8 reaction cycle then they are loss the activity in 9 to 11 cycles .the 4-(4-hydroxybenzylidene)-3-methylisoxazol-5(4H)-one structure is the confirmed by the -C=O group have the carbon NMR peak gave at 168.81ppm and C=N at 163.84 ppm in proton the 11.06 is confirm the free $CH_3$ group. and near about actual the reaction mechanic of the first free electron pair of ammonium hydroxide hydrochloride are attack on carbonyl group of ester in the presence of the silica supported aluminium hydroxide due the aluminium hydroxide have both property some reaction its act as acid catalyst as well as base they make the oxime. the oxygen is taken attack on the carbonyl carbon undergoes to H- mobility reaction with resent to remove ethanol from reaction mass. the catalyst is separate by the compound dissolve in ethanol and separate by separating funnel because the silica supported catalyst are insoluble in solvent. then they are using for crystallization procedures.

Table 1: process for solvent selection

| Entry | Catalyst | Solvent | time | Yield |
|---|---|---|---|---|
| 1 | 1 | EtoH | 90 | 85 |
| 2 | 1 | EtoH+Water | 90 | 94 |
| 3 | 1 | Acetone | 90 | 70 |
| 4 | 1 | MeOH | 90 | 72 |
| 5 | 1 | Water | 90 | 79 |
| 6 | 1 | Solvent free | 90 | 65 |

The solvent ratio is ethanol+water(1:1) is good solvent in case of this type of catalyst. For the next step is the amount of the catalyst and time, temp for the Economical as well as environmental friendly reaction path.

Table 2:

| Sr | Amount of catalyst | temperature | time | yield |
|---|---|---|---|---|
| 1 | 0.1 | RT | 90 | Ta |
| 2 | 0.1 | 45 | 90 | Ta |
| 3 | 0.1 | 60 | 90 | Ta |
| 4 | 0.1 | 75 | 90 | Ta |
| 5 | 0.1 | 90 | 90 | 12 |
| 6 | 0.2 | RT | 90 | Ta |
| 7 | 0.2 | 45 | 90 | Ta |
| 8 | 0.2 | 60 | 90 | Ta |
| 9 | 0.2 | 75 | 90 | 16 |
| 10 | 0.2 | 90 | 90 | 24 |
| 11 | 0.3 | RT | 90 | Ta |
| 12 | 0.3 | 45 | 90 | Ta |
| 13 | 0.3 | 60 | 90 | 14 |
| 14 | 0.3 | 75 | 90 | 18 |
| 15 | 0.3 | 90 | 90 | 27 |
| 16 | 0.4 | RT | 90 | Ta |
| 17 | 0.4 | 45 | 90 | 13 |
| 18 | 0.4 | 60 | 90 | 17 |
| 19 | 0.4 | 75 | 90 | 21 |
| 20 | 0.4 | 90 | 90 | 34 |
| 21 | 0.5 | RT | 90 | 12 |
| 22 | 0.5 | 45 | 90 | 27 |
| 23 | 0.5 | 60 | 90 | 34 |
| 24 | 0.5 | 75 | 90 | 39 |
| 25 | 0.5 | 90 | 90 | 96 |
| 26 | 0.6 | RT | 90 | 12 |
| 27 | 0.6 | 45 | 90 | 29 |
| 28 | 0.6 | 60 | 90 | 35 |
| 29 | 0.6 | 75 | 90 | 40 |
| 30 | 0.6 | 90 | 90 | 95 |

The above table 1 indication for the efficient temperature and amount of the catalyst for reaction,

*Ta: Trace amount, catalyst amount in mmole%

Table 3

| Der. num | Aromatic aldehyde | MP*c(Range) | | Yield(%) | |
|---|---|---|---|---|---|
| | | Practical MP | Lit MP | Practical Yield | Lit Yield |
| 1 | C6H5 | 141-143 | 140-142 | 89 | 81 |
| 2 | O-CH3- C6H5(3) | 215-216 | 216-218 | 96 | 84 |
| 3 | O-CH3- C6H5(4) | 172-174 | 173-175 | 94 | 86 |
| 4 | NO2- C6H5(2) | NF | NF | NF | NF |
| 5 | NO2- C6H5(3) | NF | NF | NF | NF |
| 6 | NO2- C6H5(4) | NF | NF | NF | NF |
| 7 | OH- C6H5(2) | 199-201 | 198-200 | 91 | 85 |
| 8 | OH- C6H5(3) | 133-134 | 134-136 | 87 | 82 |
| 9 | OH- C6H5(4) | 211-213 | 210-212 | 85 | 83 |
| 10 | Cl- C6H5(2) | NF | NF | NF | NF |
| 11 | Cl- C6H5(3) | NF | NF | NF | NF |
| 12 | Cl- C6H5(4) | NF | NF | NF | NF |
| 13 | 2-furyl | 239-240 | 238-241 | 83 | 81 |
| 14 | Br- C6H5(4) | NF | NF | NF | NF |
| 15 | OH-C6H5(3,4) | 212-214 | 211-212 | 85 | 80 |

*2,3,4 indicates the substitution of functional group, NF= Not formed

(1)4-benzylidene-3-methylisoxazol-5(4H)one:

Colour: White

IR:3365,3253,2368,1660,1625,1307,1143,1058,1006

$^{1}$HNMR: 2.28(3H),7.43(1H),7.54-7.57(2H),7.80(1H),8.45(1H)

$^{13}$CNMR: 11.69,113.71,119.21,127.12,137.45,151.27,162.21,169.44

MASS: 188.2

(2) 4-(3-methoxybenzylidene)-3-methylisoxazol-5(4H)one:

Colour: yellow

IR:3585,1715,1625,1557,1428,1192,1095,

$^{1}$HNMR:2.28(3H),3.85(3H)(O-$CH_3$),6.85-7.28(4H)(Aromatic ring),7.89(1H)

$^{13}$CNMR:19.21,58.32,121.25,125.35,129.45,134.25,150.21,161.24,169.24,172.25

MASS:217.20

(3) 4-(3-hydroxybenzylidene)-3-methylisoxazol-5(4H)one:

Colour: Yellow

IR:3425,3225,2355,1724,1561,1296,1172,665

$^1$HNMR:2.25(3H),6.94-6.97(2H),7.80(1H),8.45-8.47(2H),11.06(1H)

$^{13}$CNMR:11.27,113.81,116.13,124.54,137.51,151.52,162.26,163.84,168.81

MASS:204.54

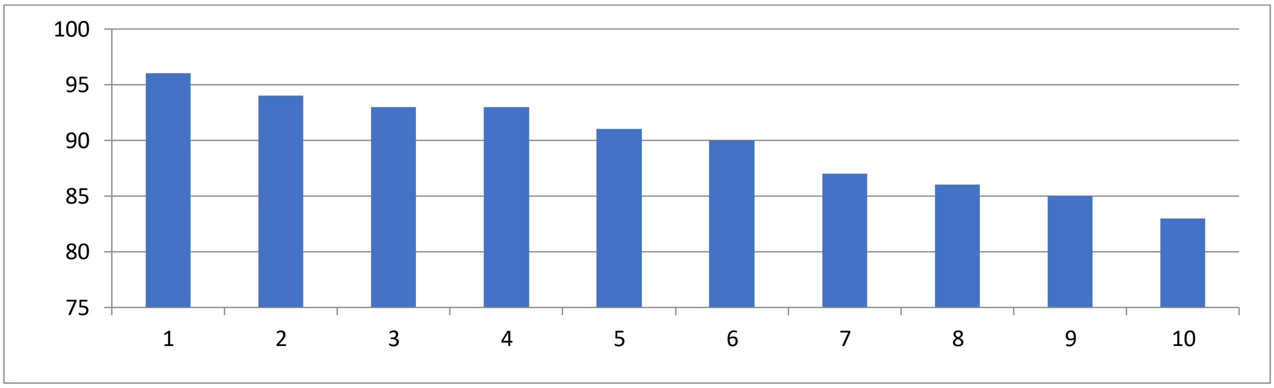



**Figure 2: Reusability cycle of catalyst**

We show the above figure:2 the cycle num 1 to 9 gave good yield after cycle 9 its decress.

Conclusion: Above table:3 is indicates the methoxy functional group is gave good yield then the hydroxy group and the nitro substituted aldehyde they does not formed as a product so we are found the electron rich group are gave good yield after the reactions and the approximately the catalyst are almost equivalent work up to 8 to 9 reaction cycles. the generally the previously used catalysts are not more than 5 times reused and its disposed out side its harmful in some cases but over catalyst is highly reusable and easy to disposed outside because aluminium hydroxide is good anti bacterial activity.

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